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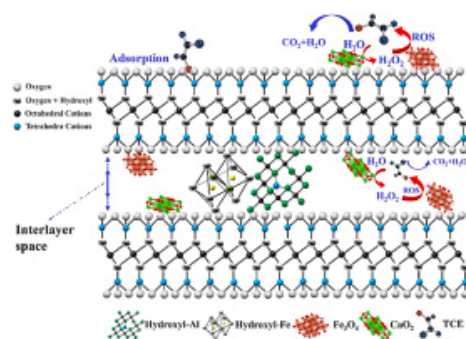
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Al-Fe pillared bentonite-Fe₃O₄-CaO₂ nanocomposite for efficient adsorption and degradation of trichloroethyleneMeiyun Feng^{a,1}, Zhiqiang Xu^{a,1}, Xue Bai^a, Kuangfei Lin^a  , Meng Zhang^b  [Show more](#)  Share  Cite<https://doi.org/10.1016/j.scp.2023.100995> [Get rights and content](#) 

Abstract

The agglomeration of magnetic nanoparticles significantly affects their catalytic efficiency, and the reduction of oxidant dosage is crucial for the practical application of heterogeneous catalysis from the perspective of cost and environmental friendliness. Al-Fe pillared bentonite-Fe₃O₄-CaO₂ nanocomposite (PB@Fe₃O₄-CaO₂) was synthesized in this study to form a heterogeneous catalytic system, and trichloroethylene (TCE) was used as the target contaminant because of its universality and volatility. The adsorption of TCE in the PB@Fe₃O₄-CaO₂ system reached 63.9% under the condition of release inhibition of H₂O₂ from CaO₂, and the value decreased to 28.2% and 29.8% under conditions favoring H₂O₂ release owing to the efficient catalytic degradation of TCE (68.0% and 65.4%, respectively). A higher systematic pH would result in a significant decrease in the TCE removal efficiency in the PB@Fe₃O₄-CaO₂ system owing to the decrease in the yield of H₂O₂ and the tendency toward O₂ generation. The co-existing inorganic anions and cations also resulted in a decrease in the efficiency of TCE removal, which was attributed to the generation of weak free radicals and promote the aggregation of the catalyst, respectively. Although the adsorption effect was 50.9% for TCE removal in actual groundwater, the degradation efficiency of TCE in the PB@Fe₃O₄-CaO₂ system (41.9%) was approximately the same as that of the PB@Fe₃O₄/H₂O₂ system (40.9%) considering TCE volatilization. This research provides a material for the efficient adsorption and degradation of trichloroethylene without the need for the exogenous addition of oxidants.

Graphical abstract



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Introduction

Advanced oxidation processes (AOPs) have been extensively studied for wastewater treatment (Deng and Zhao, 2015), sludge dewatering, micropollutant removal (Lin et al., 2022), the removal of gaseous volatile organic compounds (Dai et al., 2021), and the remediation of organic contaminated soil (Zhou et al., 2019) since their proposal in the 1980s. Compared with homogeneous activated approaches for the generation of strongly oxidizing species (SOS) based on H_2O_2 , HSO_5^- , and $\text{S}_2\text{O}_8^{2-}$, including transition metal ion activation, alkali activation, thermal activation, ultraviolet activation, and a heterogeneous catalytic system (HCS), characterized by a wider operating pH, energy conservation, and short reaction time (Garcia-Costa and Casas, 2022; Pai and Wang, 2022; Tian et al., 2021). Most research on HCS focuses on the performance of catalysts, including the efficiency-affecting factors, removal mechanism, recycling, and reusing ability (Huang and Zhang, 2019; Kurian, 2021; Huang et al., 2021; Ding et al., 2022).

In HCS, oxides are commonly used as supports to disperse active metals and to alleviate their sintering and deactivation (Védrine, 2019), and electron transfer between metal oxides and peroxides occurs at the interface to produce SOS (Ji et al., 2022). Fe_3O_4 magnetic nanoparticles (NPs) that possess intrinsic peroxidase-like activity have drawn wide attention and can degrade organic substrates through heterogeneous Fenton-like reactions (Gao et al., 2007; Munoz et al., 2015). To avoid the aggregation of Fe_3O_4 NPs, owing to their magnetic properties and improved catalytic activity, researchers have widely studied the use of carbon materials as supports for Fe_3O_4 NPs immobilization (Zhang et al., 2013; Zubir et al., 2014; Xu et al., 2016). Bentonite has been used as a support for Fe_3O_4 NPs for the adsorption and degradation of rhodamine B (Wan et al., 2017) and brilliant blue (Ain et al., 2020) and for the treatment of stabilized landfill leachate (Ma et al., 2018) through heterogeneous Fenton-like processes.

The catalytic process always occurs on the surface of the catalyst in HCS (Xu and Wang, 2012). However, the mass transfer of H_2O_2 from the solution to the catalyst surface is difficult because of its high hydrophilicity and instability property of H_2O_2 (Yang et al., 2018). The bulk feeding of H_2O_2 in HCSs may also result in potential safety hazards, a high cost, and excess consumption of chemicals, similar to homogeneous catalysis (Asghar et al., 2015). The adsorption of organic substances on HCS is a prerequisite for their degradation and is facilitated by the large specific surface area of the catalysts. However, an excessively high surface loading of contaminants can lead to incomplete degradation (Lou et al., 2014; Jafari et al., 2017). Although most of the

reactions can be completed rapidly owing to the large number of free radicals generated instantaneously with a large amount of oxidant (Liu et al., 2021), the self-quenching reaction of radicals occurs simultaneously, resulting in a low efficiency of oxidant utilization (Ain et al., 2020). In addition, incomplete degradation or mineralization of target pollutants occurs when a finite oxidant is used (Li et al., 2014). Therefore, degrading all the adsorbed pollutants with an exact amount of oxidant is difficult, and excess catalyst can increase costs and environmental risks.

In some cases, the application of in situ-generated H₂O₂ is potentially safer and more economical than bulk commercial H₂O₂ (Yalfani et al., 2011). The ability of formic acid, hydrazine, and hydroxylamine to act as H₂ substitutes for in situ H₂O₂ generation is investigated during phenol degradation in the Fenton reaction (Yalfani et al., 2011). A Zn⁰-CNTs-Fe₃O₄ composite that can in situ generate H₂O₂ and radical •OH with dissolved oxygen was investigated for the degradation of 4-chlorophenol (Yang et al., 2018). In addition to the in situ generations of H₂O₂ based on substrate reactions, oleylamine-coated CaO₂/Fe₃O₄ nanocomposites can realize the oxygen-independent generation of ROS and highly efficient cancer treatment. CaO₂ reacts with water to generate H₂O₂, which can be catalyzed by Fe₃O₄ to form hydroxyl radicals (•OH) to inhibit tumor growth (Mamat et al., 2021). In this study, Al-Fe pillared bentonite-Fe₃O₄-CaO₂ nanocomposite (PB@Fe₃O₄-CaO₂) was synthesized to form an HCS based on a controlled release material (O'Connor et al., 2018), which can catalyze the H₂O₂ that generate situ by CaO₂ through its carrier (PB@Fe₃O₄). Trichloroethylene (TCE), a typical volatile organic pollutant in the soil and groundwater of contaminated sites, was used as the target contaminant to verify the effectiveness of the system. The aim of this study was to verify the adsorption and catalytic degradation efficiency of PB@Fe₃O₄-CaO₂ for TCE, the main factors that would influence the reaction, and the remediation efficiency of TCE-polluted groundwater. The research results provide an efficient material for the remediation of volatile organic pollutants in contaminated sites and features that do not require the exogenous addition of oxidants.

Section snippets

Chemicals and reagents

Bentonite BT (3000 mesh) was purchased from Shanghai Macklin Biochemical Co., Ltd. (Shanghai, China). The details of the other materials and chemicals used in this research are provided in Supplementary Material (Text S1). All chemical reagents were of analytical grade, and ultrapure water was used to prepare experimental solutions....

Synthesis of Al-Fe pillared bentonite (PB)

The synthesis method of PB was based on a method in the literature (Barakan and Aghazadeh, 2019). In detail, 45 mL AlCl₃·6H₂O solution (0.4M) was added to the 5 mL...

Characterization of materials

SEM images of BT, PB, PB@CaO₂, PB@Fe₃O₄, and PB@Fe₃O₄-CaO₂ are shown in Fig. 1. A clear lamellar structure of BT and PB (Fig. 1 a and 1 b) was observed, and the synthesized PB@CaO₂ was characterized by a

homogeneous distribution of CaO₂ NPs on the interlayer of PB (Fig. 1 c). As shown in Fig. 1 d, the Fe₃O₄ NPs were spherical and irregularly accumulated on PB. However, the in situ synthesized CaO₂ filled the gap between Fe₃O₄ NPs, which could cause the accumulation pattern of the composites to...

Conclusions

In this study, Al-Fe pillared bentonite-Fe₃O₄-CaO₂ nanocomposite (PB@Fe₃O₄-CaO₂) was synthesized with the aim to form the heterogeneous catalysis system, which can achieve the catalyze of H₂O₂ that generate in-situ by CaO₂ through the PB@Fe₃O₄. As a typical volatile organic pollutant in the soil and groundwater of contaminated sites, TCE was chosen as the targeted contaminant to verify the effectiveness of the system. The characterization results of the materials show that the interlayer...

Credit Author Statement

Meiyun Feng: Software, Investigation, Data curation, Writing – original draft. Zhiqiang Xu: Investigation, Validation, Data curation, Writing – original draft. Xue Bai: Investigation, Software. Kuangfei Lin: Conceptualization, Writing – review & editing, Supervision, Validation. Meng Zhang: Conceptualization, Resources, Validation, Supervision, Project administration....

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper....

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